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Coherent control and high-precision spectroscopy with an optical frequency comb

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Summary

The invention of the laser has provided scientists with an excellent tool to measure and control atomic and molecular systems. In the early days of laser physics, cw and pulsed lasers were used for different types of applications. Using a single-mode laser, scientists were able to measure the absolute frequency of atomic and molecular transitions with extreme precision. This has led to the development of state-of-the-art atomic clocks and stringent tests of fundamental theories. On the other hand, so called mode-locked lasers provide a broad spectral range and a short pulse duration, which can be used to achieve a high temporal resolution. Together with pulse shaping techniques, broadband pulses can be used to control and manipulate excitation of atoms and molecules through quantum interference effects. At the turn of the 21st century, a new type of mode-locked laser was invented that brought these two worlds together. Named after its spectral properties, it is called a frequency comb laser. It emits a spectrum of equidistant modes (like a comb), which interfere such that the intensity of the output looks like an infinite train of regular ultrashort pulses. In this way one can combine the properties of a broad bandwidth of ultrashort pulses with high spectral resolution.

In the work presented in this thesis we make use of the unique properties of the frequency comb laser to control and manipulate the excitation of atomic transitions. Our method involves the excitation of atomic transitions in a counterpropagating geometry with shaped comb pulses. The broad bandwidth of these pulses allows us to excite the transition via multiple quantum pathways, and the control over the shape of the pulses via the spectral phase enables the manipulation of the quantum interference between the various pathways. Exciting these transitions in a counterpropagating geometry not only results in high-precision spectroscopy measurements, but also enables high-resolution control over resonant and nonresonant pathways. A more detailed motivation for our work is presented in Chapter 1.

Chapter 2 provides a solid theoretical background of the methods used in this thesis. We start with the propagation of electromagnetic waves in vacuum and linear media, and derive how ultrashort laser pulses are affected by propagating through dispersive materials. We continue with other pulse shaping techniques and derive the temporal and spectral properties of shaped pulses when applying sinusoidal and a V-shaped spectral phase masks. The discussion is then extended from single pulses to pulse trains and we discuss how the

coherent addition of ultrashort pulses results in a comb of equidistant narrow modes. This theory chapter is concluded with a mathematical description of light-matter interaction for two and three level atoms and a discussion of the differences between resonant and nonresonant two-photon absorption (TPA). The main experimental result of this thesis is the manipulation of two-photon signal when excited by counterpropagating frequency comb pulses. To predict the influence of pulse shaping on the two-photon signal we use the analogy between excitation in a copropagating and counterpropagating geometries. In Chapter 3 we investigate the effect of pulse shaping on the signal in the copropagating geometry for resonant and nonresonant TPA. A V-shaped spectral phase mask is applied, which splits each pulse into a red and blue subpulse. We show that the time delay between the pulse pair, and the time delay between the red and blue subpulses have a strong influence on both the overall shape and the oscillation of the signal. We also discuss the contributions of photon pairs from different pulses (and subpulses) and how the interference between these pathways leads to the total signal. The insights gained from these results are used in later chapters to model the counterpropagating signal.

In Chapter 4 we use pulse shaping on counterpropagating frequency comb pulses in order to reduce the dominant Doppler-broadened background. We show that applying a V-shaped phase mask completely eliminates the background signal and therefore results in an excellent signal-to-noise ratio. The usability of this technique is illustrated by performing high-precision spectroscopy on the $5S \rightarrow 7S$ transition in Rb, with about one order of magnitude improved frequency accuracy compared to previous measurements.

In Chapter 5 we generalize the concept presented in the previous chapter to arbitrary pulse shapes. We extend the model from Chapter 3 to the counterpropagating geometry and derive an equation to predict the excitation pattern resulting from counterpropagating shaped pulses. This equation reveals a number of intriguing symmetry properties of the signal, which we experimentally demonstrate by applying a sinusoidal spectral phase mask. It is shown that small changes in the spectral phase mask lead to dramatically different excitation patterns, with excellent agreement between the experimental results and the numerical simulations.

For nonresonant TPA all pairs of frequencies that participate in the excitation have approximately the same weight in the transition amplitude. However, in resonant TPA, pairs of photons where one of the photons has a frequency near an intermediate state lead to a larger excitation amplitude. Moreover, the contribution of the resonant pathway (a pair of photons that is also resonant with the intermediate state) has a phase shift with respect to the nonresonant pathways. When excited in the counterpropagating geometry the different pathways also behave differently to the Doppler effect. In Chapter 6 we discuss these aspects and experimentally show that the frequency comb parameters can be

used to discriminate between resonant and nonresonant pathways. By applying a V-shaped spectral phase mask we show that the quantum interference between the resonant and nonresonant pathways becomes spatially dependent. In coherent control experiments the spectral phase mask can potentially have hundreds of degrees of freedom and can assume almost any shape. Searching in such a high-dimensional space is impractical and does not necessarily provide useful insights about the physical system. The phase mask is therefore usually parameterized into a function with a limited set of control parameters, such as the V-shaped phase and the sinusoidal phase used in this thesis. In Chapter 7 we extend the concept of sinusoidal phase masks to arbitrary periodic phase modulation. We show that any periodic spectral phase mask results in a sequence of subpulses where the amplitude of the subpulses depends on the coefficients of the Fourier decomposition of the periodic function. This approach provides a larger set of control parameters for searching through a high-dimensional control landscape. This is illustrated by calculating the non-resonant TPA signal with 2 and 3 control parameters where we show how adding control parameters provides a better understanding of the control landscape.